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Leaching of different elements from subbase layers of alternative aggregates in pavement constructions

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Abstract

The objective of this study was to analyze the accumulated effects of leaching in two test roads were municipal solid waste incineration (MSWI) bottom ash and aggregate from a railway embankment, respectively, were used as subbase aggregates. Solid samples from the subbase and the subgrade were collected in trenches, which were excavated perpendicular to the road extension. The samples were analyzed with respect to pH, water content, electrical conductivity and extractable fractions of macro and trace constituents. To conclude, spatial distribution patterns of different constituents in subbase and subgrade layers confirms the existence of two major transport processes in a road with permeable shoulders: diffusion underneath surface asphalt layers driven by a concentration gradient directed horizontally towards the shoulder of the road where the dissolved elements are carried away by advection.

Keywords: Transport processes; Asphalt pavement; Shoulder; Advection; Diffusion

1. Introduction

The recycling and use of alternative materials, generated from waste materials and residues, such as municipal solid waste incineration (MSWI) bottom ash, steel slag and crushed concrete, for building and construction purposes may be a useful approach to save natural resources. But at the same time the utilization of these materials may imply a risk for environmental impact due to the leaching of contaminants. The release of contaminants for different disposal and utilization scenarios is governed by the system parameters, different processes and their interactions.

A systematic description of the process system of a conventional paved road construction, presented in [1], indicated that shoulders permeable for gases and water might be of great importance for the release of contaminants. The differences in leaching behavior for materials under field scale conditions and during standardized laboratory tests have been discussed by several authors. Schreurs et al. [2] found that a standard column test could be used to predict leaching from domains dominated by advection, but may not, as pointed out by [3], predict the leaching of metals from an embankment with MSWI bottom ash under field conditions very well. The hydrology, redox conditions and aging of the material under field conditions can cause huge deviations from the emission predicted by the leaching test. Brouwer et al. [4] found poor agreement between leaching of a monolithic building material (cement-stabilized fly-ash) during a laboratory diffusion test and field conditions (a paved road). The apparent diffusion coefficient of different constituents seems to be more or less equal during field conditions. The explanation given by [4] was that the transport rate is mainly controlled by a pH front moving into the material, thus diffusion is not the major rate limiting mechanism.

During recent years it has been suggested by several authors that estimations of the release of constituents should be based on a scenario approach and not on single leaching tests (often simulating conservative scenarios) or total concentration (see [5,6]). Proposed methods to assess release scenarios are based on stepwise procedures including [5–7]:

- defining release scenario
- determine intrinsic leaching parameters

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- quantifications by modeling based on the scenario chosen and leaching parameters

Simplified release models are usually based on the dominating leaching mechanism (advection or diffusion) and results from standard leaching tests are extrapolated into a field scenario; see e.g. [8,5]. Specific field conditions are simply taken into account by introducing correction coefficients. Obviously, simplified release models may cause both under- and overestimations, which in the latter case may severely reduce the possibilities of material utilization [5]. However, more precise and less conservative estimations of release will rely on more complex models and larger amounts of parameters that have to be considered. Basic knowledge of the leaching process under field conditions is required to refine the conceptual model, i.e. the qualitative description of the process system, upon which the (mathematical-) release model is based.

The objective of this study was to analyze the accumulated effects of 15 years of leaching from subbase courses in paved road constructions with permeable shoulders. This encompasses processes under the surface asphalt layers (surface course and basecourse), outside the pavement in the shoulder of the road and the transition zone between these two regions. Solid samples from two test roads were collected in two dimensions perpendicular to the road extension and brought to the lab for analysis. MSWI bottom ash and an aggregate from a railway embankment, respectively, were used as subbase aggregates in the two roads.

2. Road hydrology and mass transport

In paved road constructions with varying infiltration capacities along its surface (surface course, shoulders, median strips) the hydrology might be quiet complex (Fig. 1).

Water can enter the road body in several ways [1]; Rainwater that falls on the road construction may infiltrate directly through the surface course or at joints, cracks and deformities in the surface course, form surface runoff and infiltrate in the shoulders or it may be stored in depressions at the surface or evaporate. It may move from the shoulders laterally into the road construction driven by a capillary potential. Water may also move into the pavement structure and the embankment directly from surrounding surface or ground water. Embanked roads generally become disconnected from the underlying groundwater system and surrounding surface waters and direct infiltration of rain remains as the only water input source to the road system [9]. The saturated hydraulic conductivity of a surface course in good condition is about 10^{-8} and 10^{-7} m/s for a deteriorated pavement according to [10].

Field measurements of the moisture content in a road structure typically show a seasonal variation (e.g. [9,11]). Data from moisture content measurements in a road in Nantes, France, during a three-year period show a significant seasonal variation in all layers of the road construction [9]. The total moisture content budget on an annual basis was about 80 mm for a 1.2 m thick road construction.

Two domains subject to different hydrological conditions can easily be identified in a paved road construction: I. The volume along the unpaved shoulders of the road structure where water infiltrates at the upper boundary of the domain and moves downward under the action of gravity and capillary forces. II. The volume underneath surface asphalt layers where normally only very limited infiltration takes place. In the latter region water movement are dominated by capillarity. The boundary between these domains is not fixed and not easily defined. The water that moves downward is spread laterally by capillarity, lateral dispersion and hydraulic gradients. The location of the boundary is therefore dependent on the water flux and the moisture conditions. In the former domain dissolved contaminants are transported out of the system by advection and dispersion whereas the transport mechanisms in the latter domain usually are diffusion dominated. Hydrodynamic dispersion and diffusion will dilute dissolved constituents and reduce the concentration of dissolved species.

When considering an asphalt pavement construction, the diffusion-controlled transport regime will probably constitute the major bulk of the road construction. Once a certain constituent goes into solution the transport rates out of the diffusion-controlled regimes are controlled by the moisture content, the boundary conditions and chemical retardation processes, in addition to the physical material parameters. The conditions at the boundary between the advection and diffusion controlled

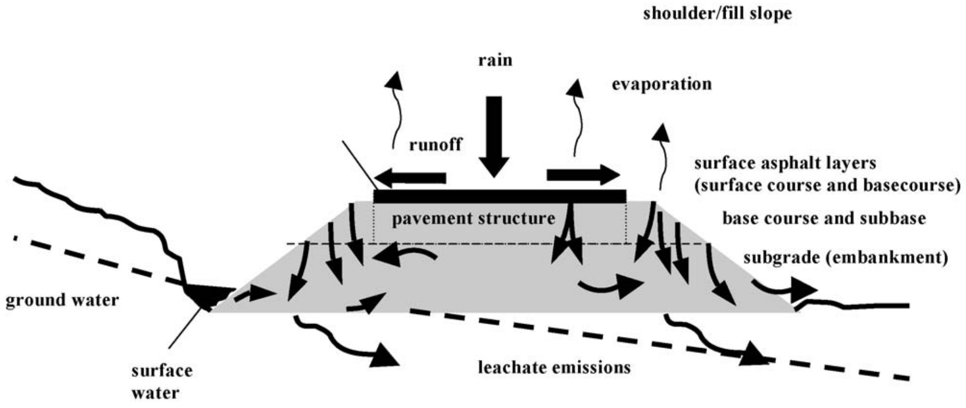

Fig. 1. Pavement construction hydrology.

regimes are influenced by the water flux in the advection controlled domain [1]. A high water flux and wash out of constituents in the advection controlled domain will gradually decrease the concentration of dissolved species in the water phase and establish concentration gradients between the advection and diffusion controlled domains.

The general experience from different field studies is that a higher degree of leaching is taking place in unpaved parts of a road construction, such as road shoulders and median strips, than within the pavement structure. Steketee [3] noticed much higher leaching of mobile substances such as chlorides from a fill slope of an embankment with aged MSWI bottom ash than underneath the pavement structure. Chloride in the slope was leached almost completely after ten years. This is in agreement with Andersson et al. [12] who found that there were significantly higher concentrations present of mobile substances in an aged MSWI bottom from a subbase layer within the pavement structure than in the road shoulder. This road construction had been in use for about 10 years. Berg et al. [13] and [14] found a certain degree of leaching of salts from a subbase of MSWI bottom ash to the subgrade below a 5 years old Danish test road. The emissions were higher compared to a reference test road constructed with natural aggregates. The results from French investigations of the subgrade underneath two paved road constructions, where MSWI bottom ash was used as subbase, indicated increased pH and electrical conductivity values in the subgrade [15]. However, the migration of heavy metals in the subgrade seems to be restricted to a few centimeters. These roads were between 20 and 22 years old at the time of the investigation. The leaching conditions are, however, a bit uncertain at both study objects since information is lacking about the road surface, especially during the first decade.

3. Study objects and sampling

This field study was carried out during 2002 in two test roads. One road (test road 1) was constructed in 1987 [16] and it was almost 15 years old at the time for this investigation. Other studies that have been performed on this object in 1998 [12] and later, in June–September 2003 [17]. MSWI (municipal solid waste incineration) bottom ash was used as subbase below an unbound base course and surface asphalt layers (Fig. 2). Table 1 shows the average chemical composition of the bottom ash in the subbase layer in 2003. The bottom ash was poorly separated and contained large pieces of incineration residues (larger than a few decimeters). Mixed shaft masses, containing mainly sand and gravel, were used as subgrade. However, some parts of the subgrade showed a higher content of fines. The whole surface course, including both roadways, was sloping towards the inner curve shoulder, which can be seen in Fig. 2. The other test road (test road 2) was built in the middle of the 1990th (Fig. 3) and was approximately 6 years at the time for this study. Aggregates from a railroad embankment were used as subbase and a till from the local area was used as subgrade. The surface course was sloping towards the shoulder.

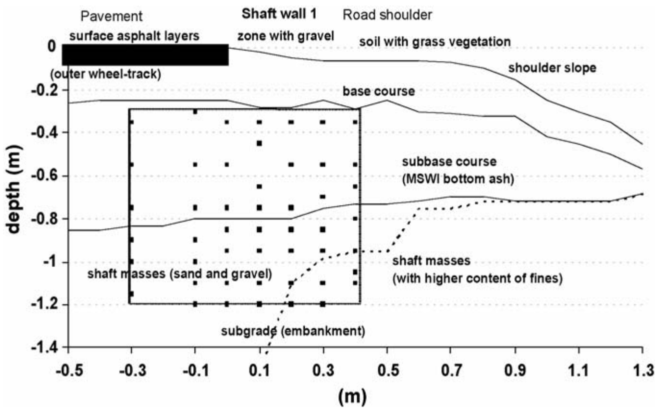

Fig. 2. Shaft wall investigated at test road 1 and the selected area. Solid samples collected are marked out by spots [18].

Table 1
Arithmetic means of the total content of Al, Ca, Fe, K, Mg, Na, Cd, Cr, Cu, Ni,Pb, S and Zn ($n = 10$) in the MSWI bottom ash (mg/kg DS) in 2003 [17]

Al	57770
Ca	60910
Fe	101910
K	16730
Mg	9652
Na	27960
Cd	10.1
Cr	240
Cu	3874
Ni	158
Pb	2688
S	2182
Zn	4541

The sampling area in each shaft wall (Figs. 2 and 3) was selected to represent domains controlled by different transport mechanisms; a diffusion-controlled domain underneath the surface asphalt layers, an advection-controlled domain in the shoulder and a transition zone beneath the edge of the asphalt cover.

Sample collection trenches were excavated into the road constructions. Each trench started in the unpaved road shoulder slope and stretched about 50 cm horizontally into the roadway (under the outer wheel-track). Solid samples (approx. 250 ml

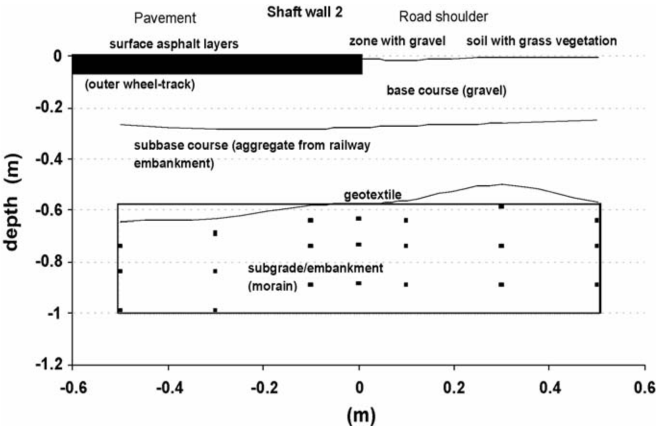

Fig. 3. Shaft wall investigated at test road 2 and the selected area. Solid samples collected are marked out by spots [18].

each) were collect from the subbase layer and the subgrade (embankment) in a check board pattern.

However, samples were only taken from the subgrade in test road 2. The wet weight of the samples varied from 300 to 450 g. Samples were stored in plastic jars until pretreatment.

4. Sample treatment and analyzing methods

All samples were treated according to a standardized procedure [19], which is a pretreatment procedure developed for soil samples before physical and chemical analysis. Each sample was dried in 40 °C. Dry material less than 2 mm was then stored and used for tests and analysis. Despite that drying at 40 °C is not a standard method for determining the gravimetrical moisture content, these results were used as an estimation of the relative difference of the moisture content between different samples.

The content of soluble salts (approximated by the electrical conductivity), pH and moisture content was analyzed for each sample. The pH of the material was measured in a soil suspension according to [20]. One volume (at least 5 ml) was mixed with 5 vol of a solution (0.01 M $CaCl_2$). The suspension was agitated for 5 min. The pH was measured after 2 h. The electrical conductivity was measured in a water solution according to [21]. The electrical conductivity gives an indication of the content of water soluble electrolytes (salts) in the material. Twenty grams of the dry material was mixed with 100 ml of de-ionized water (electrical conductivity < 0.2 mS/m) during 30 min. Each solution was filtered before the conductivity was measured. A few solutions were also analyzed to determine the easily soluble fraction of major cations (Na^+, K^+, Ca^{2+} and Mg^{2+}) and anions (Cl^- and $SO_4{}^{2-}$).

The reactive fraction of different elements was estimated by a modified extraction procedure based on the method proposed by [22]. Three grams of dry material (<2 mm) were extracted for 1 h with 60 ml 0.5 M HCl. The liquid solution/solid phase ratio (L/S) was approximately 20. The eluate was filtered by a 0.45 μm filter and stored until analysis. A ICP AES instrument P-E 3000DV optic ICP at the department of Ecology at Lund University was used to perform elemental analysis of Al, As, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, Si, V and Zn.

The extraction procedure was used to estimate an extractable and chemically available fraction of these elements. Several researchers have use extractions with dilute HCl solutions to partition “available” trace metal forms from “non-available”, resistant forms according to [22]. Sutherland [22] found that extractions of soils and road deposit sediments with 0.5 M HCl showed results similar to the total amount released during a sequential extraction procedure called the BCR (Bureau of Reference) procedure, see [23]. The constituents release during this three-step sequential extraction procedure can be divided into different fractions, the acid extractable, reducible and oxidizable phase, based on the solvent used during each step. Many secondary mineral phases (amorphous and crystalline), such as carbonates, hydroxides and oxides of Al, Fe and Mn, clay mineral and humic matter, formed during natural weathering processes are partly or fully dissolved during this extraction procedure. Our hypothesis is that most of the available parts of different elements that may participate in leaching, redistribution and enrichment processes are associated with these phases. Schreurs et al. [2] used a dissolved strong acid, 1 N HNO_3, in several extraction experiments on different drainage sand layers beneath base courses of different alternative aggregates to determine the leaching of different constituents from the base course. The method is best suited for reactive elements that are strongly bound to particles such as Co, Cu, Pb and Zn, but it may also work for less reactive and more mobile elements when transport processes are dominated by diffusion.

5. Results

The spatial distribution of the different elements within the vertical sections in the shaft walls is presented as contour maps. Kriging interpolation using default settings in Surfer 7.0 was used to calculate these maps. Variations of the moisture content, pH and electrical conductivity within these sections are presented in Fig. 4. The spatial distributions of the extractable fraction of different elements are presented in Figs. 5 and 6. The concentrations are presented as fractions of the maximum concentration values in the subbase (bottom ash) and the subgrade (Table 2) since it was not possible to identify a background level.

The average gravimetrical moisture content of the MSWI bottom ash is much higher than that of the subgrade (Fig. 4). A similar level of the moisture content in the subgrade can be found at both test roads. The moisture content of both materials is also increasing towards the depth. There is a negative gradient of the electrical conductivity towards the road shoulder in the MSWI bottom ash layer, which implies a depletion of the most soluble constituents in the shoulder. The results presented in Table 3 showing the electrical conductivity and the calculated meq/kg DS (dry bottom ash) in different parts of the road construction indicate that even bottom ash underneath the roadways is affected by this process. All the conductivity values in Table 3 were converted to meq (milliequivalent)/kg DS to facilitate the comparison between results (in μS/cm) from different test methods. A relation presented by [25] was used to convert electrical conductivity (μS/cm) into meq/l; $\sum \text{anions(meq/l)} = \sum \text{cations(meq/l)} = \text{EC(electrical conductivity)}/100$. This relation is only valid for conductivity values up to around 2000 μS/cm. Then meq/l was

Table 2
Maximum and minimum values of the extractable fraction of Ca, K, Cu, Pb and Cr (mg/kg DS) in the subbase and the subgrade (Test road 1 and 2)

Element	Test road 1				Test road 2	
	Subbase		Subgrade		Subgrade	
	Max	Min	Max	Min	Max	Min
Ca	54386	32228	17016	4621	13098	420
K	2776	1913	81.9	6.0	78.0	40.0
Cu	1809	682	27.0	0.29		
Pb					32.0	2.0
Cr	31.2	20.3	3.0	0.34	22.8	0.14

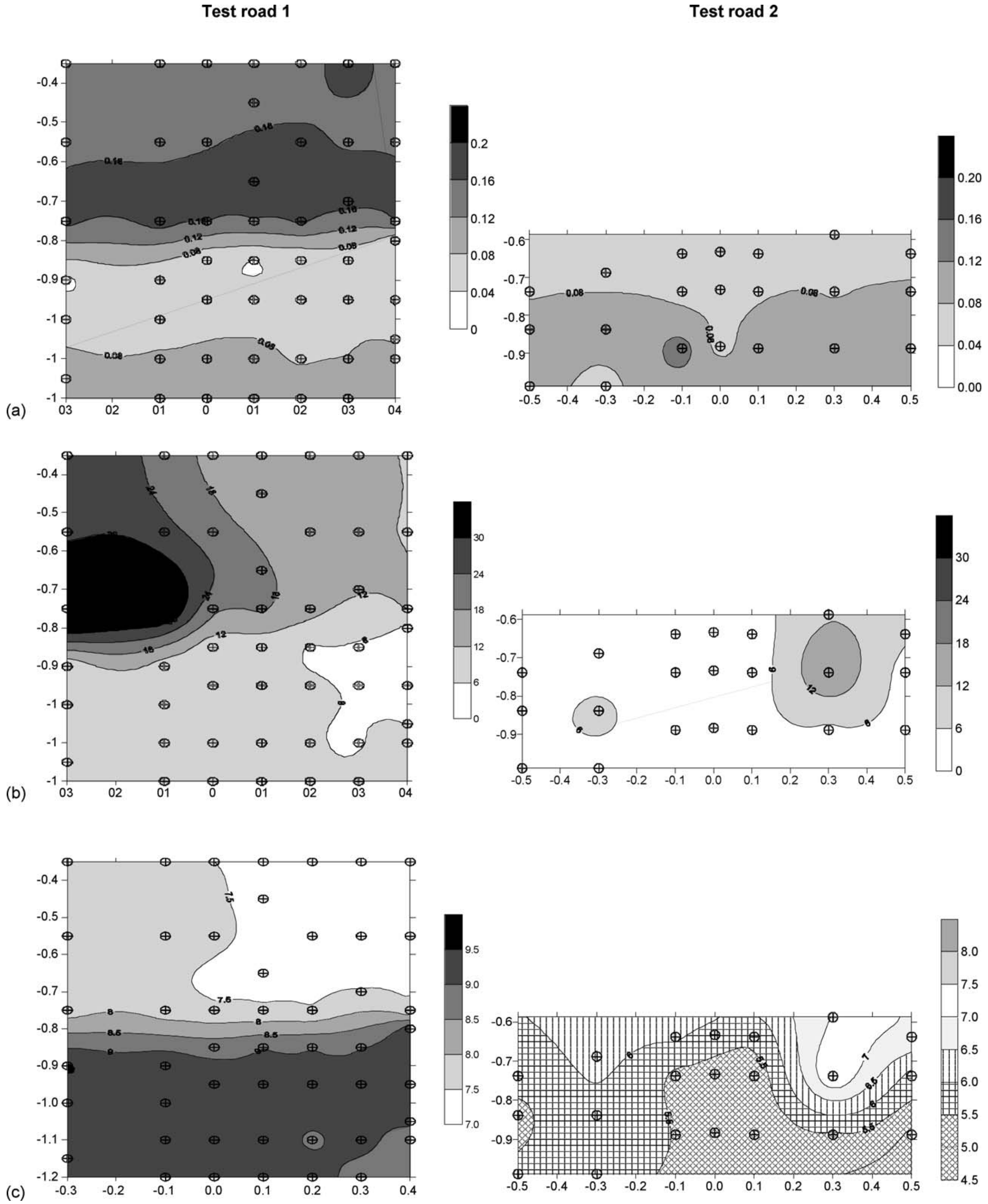

Fig. 4. Contour maps showing the spatial variations of the (a) moisture content, (b) electrical conductivity (mS/m) and (c) pH in the MSWI bottom ash and the subgrade at test road 1 (left), and in the subgrade at test road 2 (right). The edge of the asphalt cover will be found at zero on the *x*-axis. The position of solid samples collected is marked by ⊕.

converted to meq/kg DS by considering the liquid solution/solid phase ratio (L/S) defined for each test method. The contour maps of the extractable fraction of elements such as Ca (Fig. 5) and S show a distribution pattern very similar to that of the easily soluble fraction dissolved during the electrical conductivity analysis despite that the concentrations are much higher. The easily soluble fraction of sulfate varies between 12 and 126 mg/kg DS, while the extractable fraction of sulfate varies between 509 and

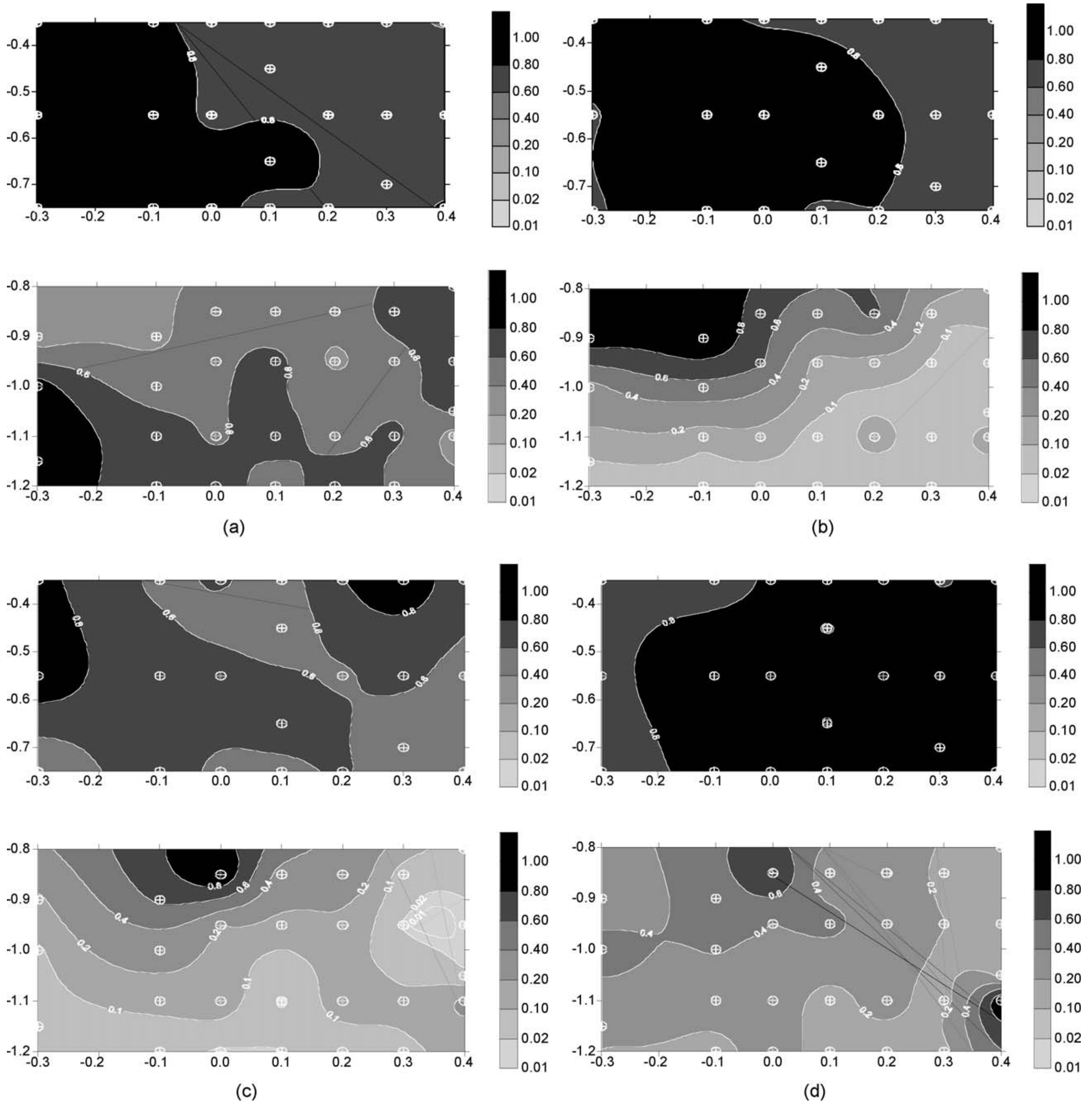

Fig. 5. Contour maps showing spatial variations of the extractable concentrations of (a) Ca, (b) K, (c) Cu and (d) Cr in the MSWI bottom ash layer (upper) and the subgrade (lower) at test road 1. The edge of the asphalt cover will be found at zero on the *x*-axis. Concentrations are presented as fractions (0.8, 0.6, 0.4, 0.2, 0.1, 0.02 and 0.01) of the maximum values measured in the bottom ash and the subgrade (Table 1). The position of solid samples collected is marked by ⊕.

1446 mg/kg DS. Many elements such as Na, Al, Si, Fe, Mg, Cr and V show a pattern similar to that of K, i.e., a maximum concentration close to the edge of the asphalt cover and negative gradients directed both towards the shoulder slope and towards the middle of the road body. The distribution patterns of heavy metals such as Ni, Pb, Cu, Zn and Cd are more random without any specific trends. The pH of the bottom ash is generally higher than that of the subgrade. The pH of the bottom ash seems to be a little bit higher within the pavement than outside it. Measured pH values in Table 4 and Fig. 4 indicate a trend similar to the one described for the easily soluble salts, i.e. decreasing values towards the shoulder slope.

These variations could be explained by a gradual carbonatization (reactions with carbon dioxide) of the bottom ash layer starting in the road shoulder.

The spatial distribution patterns of the extractable fraction of most elements in the subgrade are similar to that of K, Ca and Cr in Fig. 5. The concentrations are highest close to the contact with the bottom ash. At the bottom of the vertical cross-section (3–4 decimeters) the concentrations of elements such as Al, Fe, Mg and Cr have decreased to <20% of the maximum value, while the concentration of elements such as K, Si, Ni, Cu, Zn and Cd have been reduced to <10%. The major part of the accumulation of these elements in the subgrade is concentrated to a zone

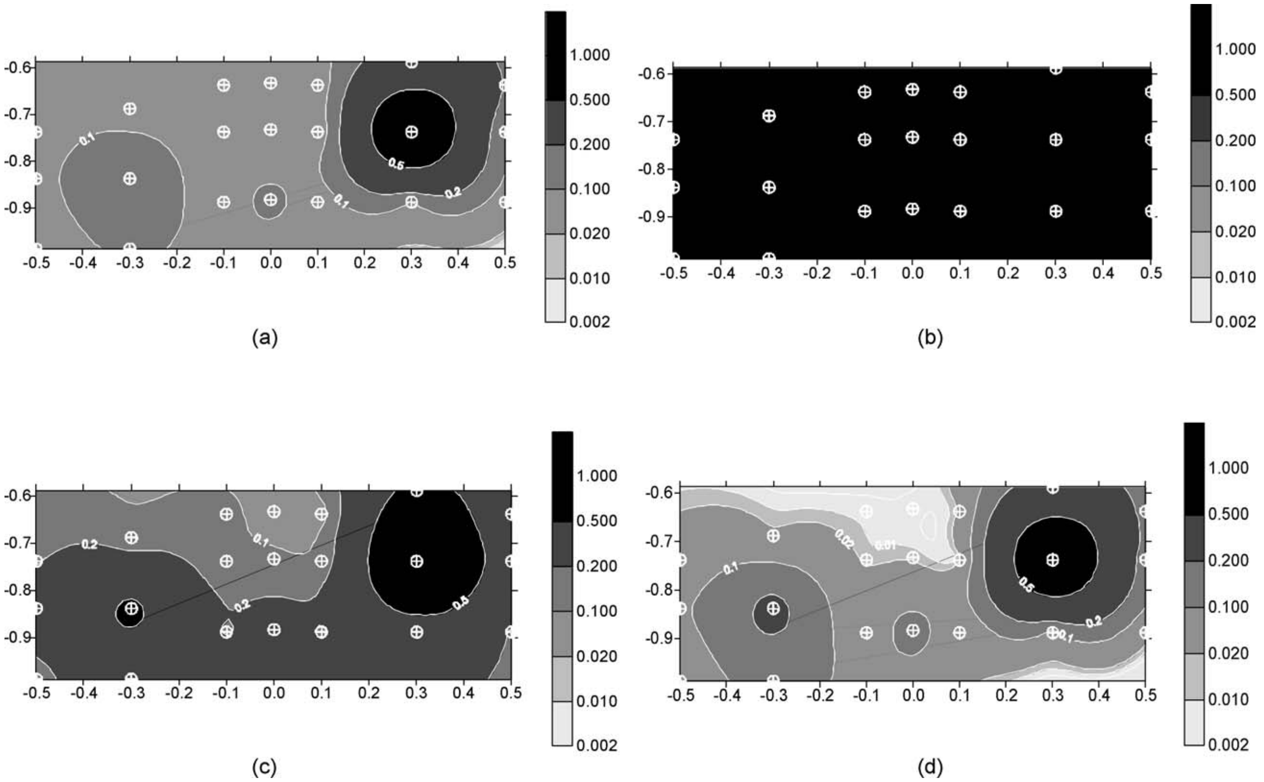

Fig. 6. Contour maps showing spatial variations of the extractable concentrations of (a) Ca, (b) K, (c) Pb and (d) Cr in the subgrade at test road 2. The edge of the asphalt cover will be found at zero on the *x*-axis. Concentrations are presented as fractions (0.5, 0.2, 0.1, 0.02, 0.01 and 0.002) of the maximum values measured in the subgrade (Table 1). The position of solid samples collected is marked by ⊕.

Table 3
The amount of easily soluble salts (in μS/cm and meq/kg DS) in different parts of test road 1

Sample position	Year	μS/cm	meq/kg	Reference
Underneath the roadway	1998	1907–2044[a]	38–41	[12]
In the shoulder	1998	300–308[a]	6	[12]
Underneath the middle of the road	2003	1250, 2300[b,c]	63, 115	
Underneath the outer wheel-track	2002	290–460[b]	14.5–23	[24]
In the shoulder	2002	<190[b]	<9.5	[24]

[a] Leaching test, L/S = 2.
[b] Determination of electrical conductivity, [19], L/S = 5.
[c] Based on samples from the same construction in 2003. Mean values (*n* = 2) from two depths, 2 and 4 dm, respectively.

Table 4
Measured pH values in different parts of test road 1

Sample position	Year	pH	Reference
Underneath the roadway	1998	8.6–8.7[a]	[12]
In the shoulder	1998	7.9[a]	[12]
Underneath the middle of the road	2003	8.93, 8.88[b,c]	
Underneath the outer wheel-track	2002	7.5–8.0[c]	[24]
In the shoulder	2002	7.0–7.5[c]	[24]

[a] Leaching test, L/S = 2.
[b] Determination of pH, [18], L/S = 5.
[c] Based on samples from the same construction in 2003. Mean values (*n* = 2) from two depths, 2 and 4 dm, respectively.

underneath the outer parts of the pavement, close to the shoulder. However, the distribution pattern of Ca (Fig. 5) and S indicate depletion instead of accumulation in the subgrade. These zones of accumulation or depletion are considered to be the result of reactions between percolating pore water contaminated by the bottom ash and the subgrade.

In the road shoulder at test road 2 a vertical accumulation zone of soluble salt (electrical conductivity) in the subgrade can be seen, in contrast to the conditions encountered at test road 1 (Fig. 4). The accumulation zone is only a few decimeters deep. There also seem to be a positive correlation between pH and the amount of soluble salts in the subgrade from test road 2. Such correlation cannot be seen in the subgrade from test road 1. The low initial pH of the subgrade in test road 2 is clearly affected by vertical percolation and mass transport. The spatial distributions of the extractable fraction of many elements (S, Ca, Si, Fe, Mg, Mn, Pb, Co, Zn, Cr and V) indicate an accumulation zone in the subgrade outside the edge of the asphalt cover (Fig. 6). However, an accumulation zone seems to be much more difficult to identify if other elements such as K are considered. The highest concentrations are found close to the contact with the subbase. Concentration contrasts between the subgrade underneath the pavement structure and in the shoulder is disappearing after a few decimeters due to decreasing concentrations in the accumulation zone and to a natural increase of the concentration of many elements towards the depth. Close to the contact between the subbase and the subgrade the difference could be more than 100 times. There are no signs of any effects on the subgrade underneath the asphalt cover.

6. Implications for modelling

For modeling purposes, the complexity of leaching under field conditions has to be reduced to something that could be handled mathematically. Still, the essence of the system characteristics must be captured. Based on a conceptual model, a mathematical model may be developed with the purpose of testing hypotheses and assumptions made in the conceptual model and, ultimately, to make predictions. The conceptual model represents a qualitative description of the system including initial and boundary conditions, state parameters, processes, and impacts at the system boundary.

It is important to stress that there is not a unique way of developing a conceptual model based on the available basic information (such as field and laboratory observations) and there is not a unique way of developing a mathematical model based on the conceptual model. The modeling uncertainty may be attributed to errors in the design of the conceptual model, uncertainty in the input parameters and errors in the numerical algorithms and calculations. The uncertainty in the input parameters may be due to limited data and knowledge and variations in time and space. Parameter variations in time are a result of slow changes of the physical and chemical properties (aging) and variations of the boundary conditions such as rain fall, temperature, mechanical impact etc. The spatial variations are due to the heterogeneity (at different spatial scales) of the material [26].

The effect of parameter uncertainty and variability is to some extent possible to quantify by sensitivity and uncertainty analysis. Uncertainty in the conceptual model is more difficult to assess but it is likely that it dominates in mathematical modelling in many cases [26].

Some important observations have been made in this study regarding the leaching process and impact of boundary conditions which ought to be taken into account in order to minimize the uncertainty in the conceptual model.

Spatial distribution patterns of different constituents in subbase and subgrade layers confirm the existence of two major transport processes in a paved road construction with permeable shoulders. The existence of diffusive transport within the pavement structure is indicated by a gradient of mobile substances in the subbase layer of MSWI bottom ash directed towards the road shoulder. This gradient was initiated and maintained by concentrated water infiltration and advective mass transport in the road shoulder. The existence of transport dominated by vertical advection at the shoulder of the road is indicated by accumulation zones of different elements in the subgrade. The concentrations of the extractable fractions in the subgrade are highest at the contact with the subbase. The accumulations are concentrated to certain zones where reactions between percolating contaminated pore water and the subgrade may have occured. No clear indications of any vertical leaching process and accumulations underneath the asphalt pavements were found.

7. Conclusion

The results presented herein are fingerprints of the accumulated effects of leaching under field conditions. The results demonstrate the complexity of the leaching process from a road construction. The spatial distribution patterns of different constituents in subbase and subgrade layers have verified the hypothesis of two major transport processes in a road construction with permeable shoulders: (i) diffusion underneath surface asphalt layers driven by a concentration gradient directed towards the shoulder of the road. (ii) transport dominated by vertical advection in the shoulder of the road. These observations have important implications for modelling of the leaching process under field conditions.

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